



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Chiharu NISHIZAWA, et al.

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For : TRANSPARENT SYNTHETIC RESIN LAMINATE
WITH PHOTOCHROMISM PROPERTY

Art Unit : 1711

D E C L A R A T I O N

Honorable Commissioner of

Patent & Trademarks

Washington, D.C. 20231

I, Kenji KOUNO, Japanese citizen, residing at c/o
Mitsubishi Gas Chemical Company Inc., Corporate Research
Laboratory, 22 Wadai, Tsukuba-shi, Ibaraki-ken 300-4247 Japan,
Declare:

That I am an inventor of the above application, and
familiar with the invention and prosecution history of said
application;

I conducted the following comparative experiments
in order to demonstrate that the transparent synthetic resin
laminate with photochromism property of the present invention

exhibits excellent photochromism property and it is impossible to form any lens by bending a transparent synthetic resin laminate obtained using CR-39 (diethyleneglycol bisallyl carbonate) as outer sheet layers;

(1) Example 1.

Example 1 is the case wherein the photochromic layer is a cured reaction product obtained from a mixture of the following reactants (A₁) and (B₁) containing (C);

(A₁): a polyurethane prepolymer obtained by reaction of diphenylmethane -4,4'-diisocyanate (X₁=4.3g) and polypropylene glycol (Y=10.7g)

(B₁): a polyurethane polyol (curing agent) obtained by reaction of diphenylmethane -4,4'-diisocyanate (X₂=2.1g) and tri-methylol propane (Z=0.9g)

(C): a tetrahydrofuran solution containing 2% of Photochromic compound 1 and 1% of Light stabilizer 1.

(2) Comparative Example 1

Comparative Example 1 is the case wherein the photochromic layer is a cured reaction product obtained from a mixture of the following reactants (A₂), (A₃) and (B₂) containing (C);

(A₂): diphenylmethane-4,4'-diisocyanate (X₁ + X₂=6.4g)

(A₃): polypropylene glycol (Y=10.7g)

(B₂): tri-methylol propane (curing agent) (Z=0.9g)

(C): a tetrahydrofuran solution containing 2% of Photochromic compound 1 and 1% of Light stabilizer 1.

(3) Comparative Example 2.

Comparative Example 2 is the case wherein the photochromic layer is a cured reaction product obtained from a mixture of the following reactants (A₁) and (B₂) containing (C);

(A₁): a polyurethane prepolymer obtained by reaction of diphenylmethane-4,4'-diisocyanate (X₁=4.3g) and polypropylene glycol (Y=10.7g)

(B₂): tri-methylol propane (curing agent) (Z=0.9g)

(C): a tetrahydrofuran solution containing 2% of Photochromic compound 1 and 1% of Light stabilizer 1.

(4) Comparative Example 3.

Comparative Example 3 is the case wherein each of out sheet layers is CR-39 (diethyleneglycol bisallyl carbonate).

Experiments

Example 1

2% of Photochromic compound 1 (1,3-dihydro-1,3,3,5,6 (1,3,3,4,5) pentamethyl-spiro[2H-indole-2,3-[3H]-naphtho[2,b] [1,4] oxazine]) to resin solid matter and 1% of Light stabilizer 1 (bis(2,2,6,6-tetramethyl-4-piperidiny)sebacate) to resin solid matter were dissolved in 8.3g of tetrahydrofuran as a solvent (C). 15g of a polyurethane prepolymer (A₁) having a NCO group equivalent weight of 1500 obtained by reaction of 4.3g of diphenylmethane-4,4'-diisocyanate and 10.7g of polypropylene glycol having an average polymerization degree of 15 and 3g of a polyurethane polyol (curing agent) (B1) having a hydroxyl group equivalent weight of 1048 obtained by reaction of 2.1g of diphenylmethane-4,4'-diisocyanate and 0.9g of tri-methylol propane were added thereto and then stirred.

The mixture solution thus obtained was coated with a doctor blade of coating thickness 400 μ m, manufactured by Yoshimitsu Seiki k.k., in Japan on a polycarbonate film of thickness 700 μ m (trade name; IUPILON, manufactured by Mitsubishi Gas Chemical Co., Inc.). After the completion of coating, the solvent was vaporized at 45°C for 10 minutes in a hot air dryer. Another polycarbonate film was further adhered to thereto in a sandwich state. Then, heat cure was performed at 70°C for 2 days. The thickness of the photochromic layer was measured. The synthetic resin laminate thus obtained was evaluated. The results were shown in Table 1.

Further, a disc of 80mm ϕ was prepared from the transparent synthetic resin laminate by punching. The disc thus

obtained was maintained at a temperature of 140°C for 5 minutes and then heat bended in a spherical mold with a curve surface (R=87mm) by press molding, whereby a lens of a spherical molded article was obtained.

Thus, the lens was normally formed without causing delamination of the photochromic layer from the disc. The appearance of the disc was very good. The result also was shown in Table 1.

Comparative Example 1

2% of Photochromic compound 1 (the same as in Example 1) and 1% of Light stabilizer 1 (the same as in Example 1) were dissolved in 8.3g of tetrahydrofuran as a solvent (C). 6.4g of diphenylmethane-4,4'-diisocyanate (A₂) and 10.7g of polypropylene glycol having an average polymerization degree of 15 (A₃) and 0.9g of tri-methylol propane (curing agent) (B₂) were added thereto and stirred.

A synthetic resin laminate was prepared from the mixture solution thus obtained in the same manner as in Example 1.

The thickness of the photochromic layer was measured. The synthetic resin laminate was evaluated. The results were shown in Table 1.

A disc was prepared from synthetic resin laminate thus obtained in the same manner as in Example 1 and heat bended by press molding in the same manner as in Example 1.

However, delamination of the photochromic layer from the disc occurred due to peeling-off of the photochromic layer from the polycarbonate films as outer layers, so that it was difficult to form a normal lens with photochromism property. The result was shown in Table 1.

Comparative Example 2

2% of Photochromic compound 1 (the same as in Example 1) and 1% of Light stabilizer 1 (the same as in Example 1) were dissolved in 0.3g of tetrahydrofuran as a solvent (C). 15g of a polyurethane prepolymer having a NCO group equivalent weight of 1500 (A₁) obtained by reaction of 4.3g of diphenylmethane-4,4'-diisocyanate and 10.7g of polypropylene glycol having an average polymerization degree of 15 and 0.9g of tri-methylol propane (curing agent) (B₂) were added thereto and stirred.

A synthetic resin laminate was prepared from the mixture solution thus obtained in the same manner as in Example 1.

The thickness of the photochromic layer was measured. The synthetic resin laminate was evaluated. The results were shown in Table 1.

A disc was prepared from the synthetic resin laminate thus obtained in the same manner as in Example 1 and heat bended by press molding in the same manner as in Example 1. However, delamination of the photochromic layer from the disc

occurred due to peeling-off of the photochromic layer from the polycarbonate films as outer layers, so that it was difficult to form a normal lens with photochromism property. The result was shown in Table 1.

Comparative Example 3

150 parts by weight of CR-39 (diethyleneglycol bisallyl carbonate) and 5 parts by weight of diisopropylperoxy carbonate as a polymerization initiator were mixed.

The liquid mixture thus obtained was poured in the hole of an O-ring with diameter 80 mm and thickness 1 mm set between two glass sheets and then polymerized and cured, whereby two sheets of a molded article were prepared. The following polymerization conditions were applied. The liquid mixture was maintained at 40°C for 4 hours and then its temperature was raised at the rate of 2.5°C/hr from 40°C to 65°C and further raised at the rate of 4°C/hr from 65°C to 85°C, Then, it was maintained at 85°C for 2 hours.

The above-mentioned mixture solution obtained in Example 1 was coated between two sheets of the molded article thus obtained to form a sandwich state and cured at 70°C for 2 days, whereby a synthetic resin laminate was obtained.

A disc was prepared from synthetic resin laminate thus obtained in the same manner as in Example 1 and heat bended by press molding in the same manner as in Example 1. However, it was impossible to bend the disc by press molding.

When bending was performed, cracking of the disc occurred, so that any lens could not be formed. The result was shown in Table 1.

Conclusion

(1) The synthetic resin laminate obtained in Example 1 exhibited excellent photochromism property of high color development speed and high color disappearance speed. Further, in Example 1, when a lens was formed from the synthetic resin laminate by the process of bending, a lens was normally formed without causing delamination of the photochromic layer interposed between two polycarbonate films of outer layers.

(2) The synthetic resin laminates obtained in Comparative Examples 1 and 2 exhibited lower color development speed and low color disappearance speed than that in Example 1. Thus, photochromism property of the synthetic resin laminates in comparative Examples 1 and 2 was inferior to that of Example 1. Further, when each lens was formed from each the synthetic resin laminate obtained in Comparative Examples 1 and 2 by the process of bending, delamination of the photochromic layer occurred due to peeling-off of the photochromic layer from outer layers. Thus, it was very difficult to form a normal lens with photochromism property from each the synthetic resin laminate obtained in Comparative Examples 1 and 2 by the process of

bending.

(3) When a lens was formed from the synthetic resin laminate obtained in Comparative Example 3 using CR-39 as outer layers by the process of bending, cracking of the disc occurred. Thus, it was impossible to form any lens from the synthetic resin laminate obtained in Comparative Example 3.

Table 1

Example & Comparative Example	Thickness of photochromic layer (μ m)	Transmittance in maximum absorption wave length		Color development speed ta (sec)	Color disappearance speed tb (sec)	Appearance	Curve bending
		Ultraviolet light					
		in non- irradiation (%)	in irradiation (%)				
		(%)					
Example 1	180	82	57	14	17	good	○
Comp.Ex.1	181	82	60	45	110	good	× occurrence of delamination
Comp.Ex.2	179	82	59	38	95	good	× occurrence of delamination
Comp.Ex.3	175	—	—	—	—	—	× occurrence of cracking

The undersigned declarant declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this day of August 18, 2004

Kenji Kouno

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